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(56) Documents Cited

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(54) Flame retardant styrene polymer

(57) A non-halogenated flame retardant polystyrenic resin, comprises 100 weight parts of styrene polymer; 1 to 40 weight parts of heat expandable graphite; 1 to 30 weight parts of red phosphorus; 1 to 30 weight parts of phosphoric ester; and 0.1 to 20 weight parts of rubber. It can be applied to various industrial substances demanding flame retardance, such as appliances, electric and/or electronic parts, packaging materials and, construction materials. It is prepared by mixing the composition by means of a mixer equipped with rotary stirrer with a speed of 500 to 5000 rpm at room temperature for a period of 1 to 10 minutes and kneading the mixture by means of a kneading extruder equipped with a screw at a speed of 200 to 300 rpm and at a temperature of 170 to 220 °C.

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NON-HALOGENATED FLAME RETARDANT POLYSTYRENIC RESIN AND
METHOD FOR PREPARING THE SAME

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The present invention relates, in general, to a flame-retardant polystyrenic resin and, more particularly, to a flame-retardant polystyrenic resin containing a synergic mixture of flame retardant agents in combination of rubber, which is superior in inflammability as well as mechanical properties and from which no harmful compounds to human body are generated upon exposing to flame. Also, the present invention is concerned with a method for preparing the non-halogenated flame retardant polystyrenic resin.

Polystyrenic resins, well-known thermoplastic resins, are useful for a plurality of industrial purposes, for example, appliances, electric and/or electronic parts, packaging materials, construction materials and the like, owing to their superior moldability. It is also known in the art that polystyrene resins, like most other synthetic resins, burn very easily and spread flames in addition to being readily molten at high temperatures. This drawback is one of the most

limitative factors that confine the fields of application of styrene polymers.

There are many methods for flame retardation of polystyrenic resins. Among them, it is the most common practice is to incorporate in styrene resin certain additives generally known as flame-retardants. Widely used are halogenated derivatives of organic compounds, halogen phosphoric ester compounds, metal hydroxides, metal oxides and the like.

Disadvantages, however, are observed in the polystyrenic resins to which these known flame-retardants or combinations of compounds are added. For example, they frequently adversely affect some of the properties of the polymer.

Metal hydroxides, inorganic flame-retardants, for example, Al(OH)_3 and Mg(OH)_2 , should be added in a large quantity to impart flame retardance to polystyrenic resins, leading to decreasing their intrinsic physical properties, especially, tensile strength and impact strength.

Brominated derivatives of organic compounds, such as decabromodiphenyloxide, tetrabromobisphenol A, tetrabromobisphenol A-bis 2,3-dibromopropylether tribromophenol, tribromophenylarylether and the like, are useful to bestow superior flame retardance upon polystyrenic resins. However, these brominated derivatives, as well familiarized, causes to gen rate tetrabromodibenzo dioxin and

tetrabromodibenzo furan, both poisonous compounds, when materials incorporating them are burning, and so European countries including Germany are taking into account that the use of them as flame retardants should be strictly restricted.

5 Also, Brominated Flame Retardant Industry Panel (BFRIP) in United states reported an examination result with regard to toxicity of brominated compounds to human body.

Other halogenated derivatives of organic compounds are of chlorine. Illustrative of the flame retardant employable in
10 polystyrenic resins are perchlorocyclodecane, perchlorocyclopentadecane, paraffin chloride and polyethylene chloride. Of these compounds, perchlorocyclodecane and perchlorocyclopentadecane are relatively poor in increasing flame retardance in the polymer because their relative low
15 molecular weight causes them to be easily transferred into the surface of the polystyrenic resin with time. Considering this problem, paraffin chloride and polyethylene chloride are more suitable to reduce or eliminate the flammability of the polymer. These chlorinated derivatives are added about twice
20 as much as brominated ones because the former is inferior in flame retardation to the latter.

Japanese Patent Laid-Open Publication No. Sho. 50-130843 discloses polystyrenic resin which is imparted with flame retardance by paraffin chloride with a chlorine content of at
25 least 70 weight percent and polyethylene chloride with a

chlorine content of 30 to 40 weight percent in combination of tribromophenylarylether. However, as previously mentioned, this brominated flame retardant, when exposed to flame, generates dioxin and furan compounds, harmful to human body.

5 In addition, it causes thermal decomposition in the course of molding the resin, thereby adversely affecting intrinsic properties thereof. Further, the gas generated when molding corrodes the molding machine.

In European Patent No. 509,535, there is described a
10 butadien-modified polystyrenic resin containing paraffin chloride, a chlorinated flame retardant, in combination with antimony trioxide and polytetrafluoroethylene. The polystyrenic resin of this patent is poor in mechanical properties because of abundance of the flame retardant (23
15 weight parts of paraffin chloride).

Apart from the above-cited patents, flame retardation for polystyrenic resin was suggested in U.S. Pat. No. 3,574,644 that teaches addition of heat expandable graphite. The resulting polystyrenic resin containing a sole flame retardant
20 of heat expandable graphite is not so good in inflammability, and the large quantity (20 weight parts) of heat expandable graphite degrades mechanical properties of the polymer.

For solving the aforementioned problems, the present inventors recognized that there exists a need for flame retardant polystyrenic resin which is superior in inflammability without a decrease of its mechanical properties and from which no harmful compounds to human body are generated when exposing to flame, and found that a polystyrenic resin containing a synergic mixture of non-halogen type flame retardants in combination with rubber satisfies the requirements.

It is therefore a principal object of the present invention to provide a non-halogenated polystyrenic resin, improved in flame retardance with its intrinsic mechanical properties maintained.

It is another object of the present invention to provide a non-halogenated flame retardant polystyrenic resin, applicable to various industrial uses with no poisonous compounds generated upon burning.

It is a further object of the present invention to provide a method for preparing the non-halogenated polystyrenic resin.

Based on the intensive and through study by the present inventors, the above objects could be accomplished by a provision of a non-halogenated flame retardant polystyrenic

resin containing a styrene polymer in combination with a synergic mixture of heat expandable graphite, red phosphorus and phosphoric ester, said resin comprising: 100 weight parts of styrene polymer; about 1 to about 40 weight parts of heat expandable graphite; about 1 to about 30 weight parts of red phosphorus; about 1 to about 30 weight parts of phosphoric ester; and about 0.1 to about 20 weight parts of rubber.

In accordance with another aspect of the present invention, there is provided a method for the preparation of non-halogenated flame retardant polystyrenic resin, comprising the steps of: mixing a composition consisting of 100 weight parts of styrene polymer, about 1 to about 40 weight parts of heat expandable graphite, about 1 to about 30 weight parts of red phosphorus, about 1 to about 30 weight parts of phosphoric ester and about 0.1 to about 20 weight parts of rubber by means of a mixer equipped with rotary stirrer with a speed of 500 to 5000 rpm at room temperature for a period of about 1 to about 10 minutes; and kneading the mixture by means of a kneading extruder equipped with a screw at a speed of 200 to 300 rpm and at a temperature of 170 to 220 degrees centigrade.

These and other objects together with others not specifically mentioned will become clear to those skilled in the art as the following description proceeds.

In accordance with the present invention, a combination of non-halogen type flame retardant agents is incorporated in polystyrenic resin. The combination of flame retardant agents consists of heat expandable graphite, red phosphorus and phosphoric ester.

Serving as the base of the non-halogenated flame retardant resin, the term "the styrene polymer" or "polystyrenic resin" as used in the specification and the appended claims, refers generally to any thermoplastic homo-
10 or copolymer composed either totally or for the most part of one or more styrene and/or vinylic compounds. For example, homopolymers made up of a monomer selected from a group consisting of styrene, alpha-methylstyrene, dimethylstyrene, ethylstyrene, chlorostyrene, bromostyrene and vinyl toluene,
15 copolymers of a combination of the monomers, and copolymers with a vinylic monomer selected form a group consisting of butadien, acrylonitrile, methylmethacrylate, isobutylene, vinyl chloride and isoprene can be useful.

Any of conventional techniques can be applied for the preparation of heat expandable graphite, and there is no particular limitation to the material graphite and process. In preparing the heat expandable graphite for the present invention, it is preferred that a graphite, such as naturally occurring crystallin graphite or heat decomposable graphite,
25 is treated with a mixture of concentrated sulfuric acid and

strong oxidant. Then, this acid treated graphite, now expandable, is washed and dried, to obtain a preferred heat expandable graphite. For example, powdery graphite with a particle size of about 20 to about 250 mesh is contacted at 5 about 45 degrees centigrade or lower with a mix solution of 98 weight percent concentrated sulfuric acid and 60 weight percent hydrogen peroxide water for a period of about 10 to about 30 minutes, washed with water and dried, to prepare heat expandable graphite. The heat expandable graphite that is 10 used mainly for graphite sheet rapidly expands to tens to hundreds times its original volume to the direction of C axis when it is exposed to flame or any source of temperature of about 500 °C or higher.

Such prepared heat expandable graphite, as it is, may be 15 used in the instant invention although preferably one which a water dispersion containing 1 weight percent of has a pH value of 4.5 to 8. This desired heat expandable graphite can be prepared by, in the course of the washing with water, contacting it with ammonia water and alkaline metal and/or 20 alkaline earth metal hydroxide in water and then, separating and drying.

From an aspect of dispersibility in polystyrene resin, the heat expandable graphite ranges in particle size preferably from about 30 to about 250 mesh and more preferably 25 from about 40 to about 200 m sh. For example, if too large

graphite, i.e. 20 to 30 mesh, is used, it is virtually impossible to obtain a polystyrenic resin with a superiority in flame retardance because of meager dispersibility, in spite of its quite high coefficient of heat expansion. On the other
5 hand, if graphite with too small particle size is used, there is a rapid decrease in heat expansion, which results in a remarkable reduction in flame retardance effect. The particle size of the heat expandable graphite can readily controlled by, for example, pulverizing the material graphite or the
10 obtained expandable graphite.

In accordance with the present invention, the heat expandable graphite is incorporated in polystyrenic resin preferably in an amount of about 1 to about 40 weight parts based on 100 weight parts of polystyrenic resin, and more
15 preferably about 2 to about 30 weight parts. For example, if too little graphite is used, there is little or no flame retardance in the resin. On the other hand, if too much graphite is used, the resulting polystyrenic resin becomes detrimental in processability and mechanical properties.

20 For a synergy effect on flame retardance in the polystyrenic resin, red phosphorus as a non-halogen type flame retardant agent is used. In accordance with the present invention, the amount of red phosphorus is preferably on the order of about 1 to about 30 weight parts based on 100 weight
25 parts of polystyrenic resin and, more preferably on the order

of about 2 to about 25 weight parts. For example, if less than 1 weight part of red phosphorus is incorporated in 100 weight parts of polystyrenic resin, there is no synergy effect on flame resistance with the heat expandable graphite. On the 5 other hand, more than 30 weight parts of red phosphorus deleteriously influences on mechanical properties of the styrene polymer.

Phosphoric ester is used in order to enhance the compatibility of the flame retardant agents with polystyrenic resin as well as to increase the synergy effect on flame retardance therein. Preferred phosphoric ester compounds of the present invention include ammonium polyphosphate, melamine phosphate, tricresyl phosphate, tributyl phosphate, triethyl phosphate, trimethyl phosphate and cresyldiphenyl phosphate. 10 In accordance with the present invention, phosphoric ester is incorporated preferably in an amount of about 1 to about 30 weight parts based on 100 weight parts of polystyrenic resin and, more preferably in an amount of about 2 to about 25 weight parts. For example, if too little phosphoric ester is 15 incorporated, no synergy effect on flame resistance is obtained in the resulting polystyrenic resin. On the other hand, if the amount of phosphoric ester is over the upper limit, there is a remarkable decrease in mechanical properties 20 of the resulting polystyrenic resin.

25 In order to prevent the styrenic resin incorporating the

above flame retardant agents therein from being degraded in mechanical properties, rubber is added to modify the styrenic resin. In accordance with the present invention, it is incorporated preferably in an amount of about 0.1 and about 20 weight parts based on 100 weight parts of polystyrenic resin and, more preferably in an amount of about 0.2 to about 15 weight parts. For example, if too little rubber is used, the effect of improving mechanical properties in the resulting polystyrenic resin is negligibly insignificant. On the other hand, if too much rubber is incorporated, the flame retardance thereof is degraded. Representative are styrene-butadien block copolymer, styrene-isoprene copolymer, styrene-ethylene-styrene block copolymer, styrene-isoprene-styrene block copolymer, and styrene-ethylene-butadien-styrene block copolymer. Most preferable is styrene-butadien-styrene block copolymer which contains a styrene content of about 20 to about 40 % by weight. If the styrene content in styrene-butadien-styrene block copolymer exceeds the upper or lower limit, compatibility with polystyrenic resin becomes poor, which results in dissipating the expecting effect of an improvement in mechanical properties.

In addition to the essential ingredients mentioned above, the non-halogenated flame retardant polystyrenic resin of the present invention, if necessary, may contain other additives, such as dispersing agents, antioxidants, uv absorbents, light-

protective agents, lubricants, antistatic agents, and organic and inorganic fillers.

In accordance with another aspect of the present invention, the non-halogenated flame retardant polystyrenic resin is prepared by mixing polystyrenic resin, the flame retardants and rubber at room temperature for minutes by means of a mixer equipped with rotary stirrer with a speed of hundreds to thousands rpm, such as ribbon blender, V-blender and Henschel mixer and then kneading the mixture at a temperature of 170 to 220 °C by means of a kneading extruder equipped with a screw rotating at a speed of 200 to 300 rpm, such as ordinary extruder, Brabender plasticorder, Banbury mixer, mixing roll or kneader.

A better understanding of the present method may be obtained in light of the following examples which are set forth to illustrate, and are not to be construed to limit the present invention.

EXAMPLE I

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100 weight parts of impact resistant polystyrene with a melt index of 4.0 g/10min. a tensile strength of 320 kg/cm², an elongation ratio of 35% and an IZOD notch resilience of 10 kg·cm/cm, 7 weight parts of heat expandable graphite, 10 weight parts of red phosphorus, 4 weight parts of ammonium

polyphosphate, and 6 weight parts of styren -butadien-styrene
5 ^{co}terblock polymer with a styrene content of 29 % by weight were
mixed by means of Henschel mixer at room temperature and at a
speed of 3000 rpm for 5 min. The resulting polystyrenic
mixture was kneaded by means of a twin screw extruder having
an L/D ratio of 13 at a temperature of 170 to 220 degrees
centigrade and at a screw speed of 250 rpm. The strands
leaving the extruder were cut to obtain pellets.

10 To determine the flammability and mechanical properties,
the pellets following dry were injection molded at a
temperature of 210 degrees centigrade under the conditions of
mold temperature 50 degrees centigrade and molding cycle 35
seconds to obtain test samples.

15 In mechanical properties test, the tensile strength and
elongation ratio were determined in accordance with ASTM D
638, on the test samples, together with the IZOD notch
resilience in accordance with ASTM D 256.

20 For flame retardant properties, the flammability was
determined in accordance with UL 94V, on the test samples
having a thickness of 3.2 mm, together with the critical
oxygen index in accordance with ASTM D 2863.

The appearance was determined by the presence of the flow
mark, which was observed with naked eyes.

25 The above test results are given as shown in Table 1
below.

EXAMPLES II THROUGH X

Test samples were prepared in a manner similar to that in Example I, except that the amounts of heat expandable graphite, red phosphorus, ammonium polyphosphate and styrene-butadien-styrene terblockcopolymer rubber were used as indicated in Table 1 below.

They were tested for the mechanical properties, the flame retardant properties and the appearance in the same manner as in Example I.

The results are given as shown in Table 1 below.

COMPARATIVE EXAMPLES I THROUGH IX

Test samples were prepared in a manner similar to that in Example I, except that ammonium polyphosphate was excluded and the amounts of heat expandable graphite, red phosphorus and styrene-butadien-styrene terblockcopolymer rubber were used as indicated in Table 2 below.

They were tested for the mechanical properties, the flame retardant properties and the appearance in the same manner as in Example I.

The results are given as shown in Table 2 below.

< Table I >

	Composition (weight parts)				Tensile strength kg./cm ²	Elongation at break (%)	IZOD notch resilience kg.cm/cm	Critical oxygen index	Flammability	Appearance
	Syrene polymer	Heat expandable graphite	Red phosphorus	Ammonium polyphosphate						
Example 1	100	7	10	4	6	284	19	8.6	28.6	V-O
Example 2	100	7	7	7	6	295	21	8.9	27.8	V-O
Example 3	100	7	5	9	6	286	26	9.6	27.6	V-O
Example 4	100	7	4	10	6	293	25	8.4	27.9	V-O
Example 5	100	10	7	4	6	306	18	9.2	27.5	V-O
Example 6	100	10	4	7	6	308	23	9.8	27.0	V-O
Example 7	100	10	9	4	6	315	16	6.8	32.8	V-O
Example 8	100	10	6	7	6	309	14	6.5	32.5	V-O
Example 9	100	10	4	9	6	314	19	7.0	31.9	V-O
Example 10	100	10	3	10	6	307	12	6.4	30.5	V-O

* The presence of flow mark in molding product : O : Some, X : No

< Table 2 >

Composition (weight parts)					Tensile strength kg./cm ²	Fringation (Z.D.I.)	Flammability	Appearance				
Styrene polymer	Ullent expandable graphite	Red phosphorus	Phosphoric ester	Name	Styrene-butadiene-styrene terblock copolymer	%	notch resilience kg. cm/cm	Critical oxygen index				
Comparative 1	100	0	0	Ammonium Polyphosphate	21	6	287	30	5.3	24.0	V-2	O
Comparative 2	100	0	5	Ammonium Polyphosphate	15	6	278	23	6.6	25.7	V-1	O
Comparative 3	100	0	8	Ammonium Polyphosphate	16	6	284	22	6.5	25.9	V-1	O
Comparative 4	100	7	5	Ammonium Polyphosphate	9	0	320	17	5.8	30.8	V-0	O
Comparative 5	100	7	5	Ammonium Polyphosphate	9	2	307	20	6.8	30.4	V-0	O
Comparative 6	100	7	5	Ammonium Polyphosphate	9	10	285	27	10.4	27.7	V-0	O
Comparative 7	100	7	5	Melamine Polyphosphate	9	6	276	23	8.5	24.8	V-1	O
Comparative 8	100	7	5	Tricresyl Polyphosphate	9	6	268	23	9.1	25.2	V-1	X
Comparative 9	100	7	5	Triethyl Polyphosphate	9	6	273	26	9.3	25.3	V-1	X

* The presence of flow mark in molding product : O : Some, X : No

As apparent from the examples and tables, the flame retardant polystyrenic resins according to the present invention show sufficient flame retardance even though their low contents of non-halogen type flame retardants and exhibit 5 a remarkable improvement in elongation ratio and impact strength by virtue of modification with rubber.

The present flame retardant polystyrenic resins is extensively applicable to various industrial substances demanding flame retardance, such as appliances, electric 10 and/or electronic parts, packaging materials, construction materials and the like.

Other features, advantages and embodiments of the present invention disclosed herein will be readily apparent to those exercising ordinary skill after reading the foregoing 15 disclosures. In this regard, while specific embodiments of the invention have been described in considerable detail, variations and modifications of these embodiments can be effected without departing from the spirit and scope of the invention as described and claimed.

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CLAIMS

1. A non-halogenated flame retardant polystyrenic resin containing a styrene polymer in combination with a synergic mixture of heat expandable graphite, red phosphorus and phosphoric ester, said resin comprising:

100 weight parts of styrene polymer;

1 to 40 weight parts of heat expandable graphite;

1 to 30 weight parts of red phosphorus;

1 to 30 weight parts of phosphoric ester; and

0.1 to 20 weight parts of rubber.

2. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 1, wherein said phosphoric ester is selected from a group consisting of ammonium polyphosphate, melamine phosphate, tricresyl phosphate, tributyl phosphate, triethyl phosphate, triphenyl phosphate, cresyldiphenyl phosphate and mixtures thereof.

3. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 1, wherein said rubber is selected from a group consisting of styrene-butadiene blockcopolymers, styrene-isoprene copolymers, styrene-butadiene-styrene block copolymer,

styrene-isoprene-styrene block copolymer and styrene-ethylene-butadiene-styrene block copolymer.

4. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 1, wherein said rubber is a styrene-butadiene-styrene terblockcopolymer with a styrene content of about 20 to 40 % by weight.

5. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 1, wherein said styrene polymer is a homopolymer made up of a monomer selected from a group consisting of styrene, alpha-methylstyrene, dimethylstyrene, ethylstyrene, chlorostyrene, bromostyrene and vinyl toluene, a copolymer of a combination of said monomers, or a copolymer of said monomer and a vinylic monomer selected from a group consisting of butadiene, acrylonitrile, methylmethacrylate, isobutylene, vinyl chloride and isoprene.

6. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 1, wherein said heat expandable graphite is one which is prepared by contacting a graphite having a particle size of 20 to 250 mesh with a mixed solution of 98 weight percent concentrated sulfuric acid and 50 weight percent hydrogen peroxide water at a temperature of 45 degrees

centigrade or lower for a period of 10 to 30 minutes, washing the resulting graphite with water and drying it.

7. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 6, wherein said particle size of graphite is 40 to 200 mesh.

8. A non-halogenated flame retardant polystyrenic resin in accordance with Claim 1, further comprising one or more additives selected from the group consisting of an antioxidant, a uv absorbent, a light-protective agent, a lubricant, an antistatic agent, and an organic and/or inorganic filler..

9. A method for the preparation of non-halogenated flame retardant polystyrenic resin, comprising the steps of:

mixing a composition consisting of 100 weight parts of styrene polymer, 1 to 40 weight parts of heat expandable graphite, 1 to 30 weight parts of red phosphorus, 1 to 30 weight parts of phosphoric ester and 0.1 to 20 weight parts of rubber by means of a mixer equipped with rotary stirrer with a speed of 500 to 5000 rpm at room temperature for a period of 1 to 10 minutes; and

kneading the mixture by means of a kneading extruder equipped with a screw at a speed of 200 to 300 rpm and at a temperature of 170 to 220°C.

10. A method in accordance with Claim 9, wherein said mixer is a ribbon blender, a V-blender or a Henschel mixer.

11. A method in accordance with Claim 9, wherein said kneading extruder is an ordinary extruder, a Brabender plasticorder, a banbury mixer, a mixing roll or a kneader.

12. A non-halogenated flame retardant polystyrenic resin substantially as described herein.

13. A method for the preparation of a non-halogenated flame retardant polystyrenic resin substantially as described herein.



Patent
Office

Application N : GB 9509790.3
Claims searched: 1 to 13

Examiner: Miss M. M. Kelman
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Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.N): C3K KEB KEF KEZ

Int Cl (Ed.6): C08K 3/00 3/02 3/04 3/32 13/00 13/02

Other: ONLINE:MATERIAL, PATENTS

Documents considered to be relevant:

Category	Identity of document and relevant passage	Relevant to claims
A	EP 0533102 A1 Albright and Wilson	
A	EP 0523790 A1 ECP Enichem Polimeri	
A	EP 0302987 A1 Dr. Wolman GmbH	

X Document indicating lack of novelty or inventive step
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